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# **Derivation of an Applied Nonlinear Schrödinger Equation**

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# Derivation of an Applied Nonlinear Schrödinger Equation

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## Abstract

We derive from first principles a mathematical physics model useful for understanding nonlinear optical propagation (including filamentation). All assumptions necessary for the development are clearly explained. We include the Kerr effect, Raman scattering, and ionization (as well as linear and nonlinear shock, diffraction and dispersion). We explain the phenomenological sub-models and each assumption required to arrive at a complete and consistent theoretical description. The development includes the relationship between shock and ionization and demonstrates why inclusion of Drude model impedance effects alters the nature of the shock operator.



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# 1 Introduction

Mathematical modeling of intense optical pulse propagation physics via nonlinear Schrödinger equations finds application in a very large number of fields. Optical parametric amplifiers and oscillators [23, 11], second harmonic generation [19] and third harmonic generation [36], Raman scattering [37], optical bistability [21], and solitons [24, 25, 33], are but a few of the physical phenomena with broad practical application. The scientific and engineering communities have significant efforts in the numerical modeling [29, 12, 1, 2, 13] of these phenomena for both commercial and scientific research purposes. In this report we derive a general nonlinear Schrödinger equation from first principles paying particular attention to the physical and mathematical assumptions necessary to arrive at each step. A variety of models and unit systems are used in the literature to describe various subsets of known physical effects (see Brabec [9], Couairon [13], or Zozulya [40]). Here we bring the physical phenomena of interest under the same framework using a single common notation and unit system.

The development proceeds as follows. We derive the physics model beginning with Maxwell's equations in section 2. The nonlinear and linear portions of a basic propagation model are introduced in section 2.1. We also discuss removal of the optical fast phase component resulting in an envelope propagation model and introduce a retarded coordinate system traveling at the pulse group velocity in order to simplify the description. Section 2.2 explicitly details specific nonlinear material models for the physics of interest. The instantaneous or Kerr material response is described in section 2.2.1. Section 2.2.2 describes delayed Kerr or Raman scattering and shows how and why it is generally combined with the instantaneous Kerr response in mathematical models. Ionization is discussed in section 2.2.3. The description of ionization covers multi-photon and tunneling regimes as well as recombination and saturation effects. It also includes both a simple and a Drude (ion-molecule collision) ionization current model. The latter leading to the concept of ionization current impedance. Of particular note is the development of the shock operator when including Drude model (collision) effects in the description of ionization in section 2.2.3. Inclusion of these effects, in the manner shown in our paper, leads to a difference in the form of the operator from some reports in the literature (see [13] for example). The entire mathematical model is summarized concisely in section 3. Appendix A gives an exhaustive description of all symbols used in the text.

## 2 Mathematical Physics Development

In this section we lay out the basis of the mathematical physics model. Beginning with the fundamental governing equations for electromagnetics, we develop a parabolic wave equation together with a set of nonlinear operators. The model accounts for diffraction, linear material dispersion, self-focusing (Kerr effect), ionization (energy loss and defocusing), and Raman interaction. We retain terms accounting for space-time focusing (see Zozulya [39]) and shock (see Brabec [8]). For clarity in the developments that follow we use bold-face to indicate vector variables, plain type for scalar quantities, and eventually a calligraphic font for variables describing envelope quantities (see Eqs. (29) and (30)). In general these variables are functions of time and space. The temporal Fourier transform of a quantity is a function of frequency and space and is indicated via a small caret or hat over the variable (see Eqs. (17a) and (17b)).

Classical free-space electromagnetic phenomena are governed by Maxwell's equations

$$\nabla \times \mathbf{E} = -\partial_t \mathbf{B} \quad \text{Faraday's Law} \quad (1)$$

$$\nabla \times \mathbf{H} = \mathbf{J} + \partial_t \mathbf{D} \quad \text{Ampere's Law} \quad (2)$$

$$\nabla \cdot \mathbf{D} = \rho \quad \text{Electric Gauss' Law} \quad (3)$$

$$\nabla \cdot \mathbf{B} = 0 \quad \text{Magnetic Gauss' Law} \quad (4)$$

where

$\mathbf{E}$  = Electric field strength (volts/meter)

$\mathbf{H}$  = Magnetic field strength (amperes/meter)

$\mathbf{D}$  = Electric flux density (coulombs/square meter)

$\mathbf{B}$  = Magnetic flux density (webers/square meter)

$\mathbf{J}$  = Current density (amperes/square meter)

$\rho$  = Electric charge density (coulombs/cubic meter)

are functions of time and space. In order to obtain a solution for a particular problem we need additional equations relating the field quantities. These constitutive expressions specify the relationships between field quantities that are functions of the specific material interacting with the fields. In free space we have

$$\mathbf{D} = \epsilon_o \mathbf{E} \quad (5)$$

$$\mathbf{B} = \mu_o \mathbf{H}, \quad (6)$$

where the free-space permittivity and permeability are

$$\epsilon_o = 8.854 \times 10^{-12} \text{ F/m} \quad (7)$$

$$\mu_o = 4\pi \times 10^{-7} \text{ H/m}. \quad (8)$$

### 2.1 Propagation Model

If the field interacts with a physical material, we must introduce descriptive models of the relationships between electric field and electric flux density and between magnetic field and magnetic flux density. For a dielectric material we may modify Eq. (5) by modeling the polarization response of the material. The field quantity  $\mathbf{D}$  is often given in units of Coulombs per square meter. However, it may be more intuitively considered in units of charge separation per unit volume or Coulomb meters per cubic meter. We may add a term (also with units of charge separation per unit volume) representing the total polarization response of the material

$$\mathbf{D} = \epsilon_o \mathbf{E} + \mathbf{P} = \epsilon_o \mathbf{E} + \mathbf{P}_l + \mathbf{P}_{nl} \quad (9)$$

where we have written the total polarization  $\mathbf{P}$  as a sum of *linear* and *nonlinear* responses  $\mathbf{P}_l$  and  $\mathbf{P}_{nl}$  respectively. We model the linear portion of the response as directly proportional to the electric field



amplitude. The *linear susceptibility* for a particular material tells us how much charge separation per unit volume is generated through material polarization for a given electric field strength. The linear portion of the model is generally given in the frequency domain as

$$\mathbf{D}_l(\omega) = \epsilon_o \mathbf{E}(\omega) + \epsilon_o \chi^{(1)}(\omega) \mathbf{E}(\omega) = \epsilon_o \epsilon_r(\omega) \mathbf{E}(\omega) \quad (10)$$

where the linear susceptibility  $\chi^{(1)}(\omega)$  and relative permittivity  $\epsilon_r(\omega)$  are functions of frequency, representing *linear dispersion*. In the time domain, this relationship is expressed via convolution as

$$\mathbf{D}(t) = \int_{-\infty}^t \epsilon(t-t') \mathbf{E}(t') dt' + \mathbf{P}_{nl}, \quad (11)$$

where *total linear permittivity* is given as  $\epsilon(t) = \epsilon_o \epsilon_r(t)$ .

Eqs. (1) and (2) yield the most general form of the wave equation for nonlinear optics

$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{\epsilon_o c^2} \partial_t^2 \mathbf{D}(t) + \frac{1}{\epsilon_o c^2} \partial_t \mathbf{J} = 0 \quad (12)$$

where  $\mu_0 = 1/\epsilon_o c^2$ . For our purposes, the electric flux density is then sufficiently described as in Eq. (11).

For any vector  $\mathbf{A}$  [16]

$$\nabla \times \nabla \times \mathbf{A} = \nabla(\nabla \cdot \mathbf{A}) - \nabla^2 \mathbf{A}, \quad (13)$$

which may be used in Eq. (12) to arrive at

$$-\nabla^2 \mathbf{E} + \frac{1}{\epsilon_o c^2} \partial_t^2 \mathbf{D}(t) + \frac{1}{\epsilon_o c^2} \partial_t \mathbf{J} = 0, \quad (14)$$

provided  $\nabla(\nabla \cdot \mathbf{E}) \approx 0$ . For linear, isotropic source-free media, Eq. (3) would allow us to conclude that the divergence of the electric field is identically zero. However, for nonlinear optics this is never really the case due to the more general relationship between the electric field and the electric flux density given in Eq. (11) [7]. The assumption is, however, valid in circumstances where the fields are primarily transverse (it breaks down for high numerical apertures). In the remainder of this paper we take Eq. (14), often referred to as the *paraxial* approximation, as sufficient for our purposes. Using Eqs. (11) through (14) we may write

$$\nabla^2 \mathbf{E} - \frac{1}{c^2} \partial_t^2 \int_{-\infty}^t \epsilon_r(t-t') \mathbf{E}(t') dt' = \frac{1}{\epsilon_o c^2} (\partial_t^2 \mathbf{P}_{nl} + \partial_t \mathbf{J}) \quad (15)$$

which forms the basis for development of many practical nonlinear propagation models.

If ionization is not included (current  $\mathbf{J}$  is zero in Eq. (15)), we have Brabec's [9] initial equation. If the response of the material and the electric field are transverse and the polarization may be considered linear, we have the scalar equation

$$\nabla^2 E - \frac{1}{c^2} \partial_t^2 \int_{-\infty}^t \epsilon_r(t-t') E(t') dt' = \frac{1}{\epsilon_o c^2} \partial_t^2 N_p E. \quad (16)$$

For convenience in Eq. (16) we have combined the polarization and current terms. The difference in the order of the derivatives operating on each means that we express the combined nonlinear term  $N_p$  as the sum of nonlinear polarization and the integral of current density. Although we do not explicitly state it here, the nonlinear operator is understood to be a function of the electric field. In order to create a tractable model we make a few more assumptions that allow a physical description of how the pulse *envelope* (rather than the instantaneous electric field amplitude) propagates.

In the subsequent development we make use of the Fourier transform. The many different representations of the transform differ by scale factors and conjugation. In order to provide clarity in what follows we define

the transform explicitly as

$$f(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \hat{f}(\omega) e^{-i\omega t} \quad (\text{Synthesis}) \quad (17a)$$

$$\hat{f}(\omega) = \int_{-\infty}^{\infty} dt f(t) e^{i\omega t} \quad (\text{Analysis}) \quad (17b)$$

It is important to understand the nature of the approximations made before developing the numerical model. It is useful in the following to write the Laplacian as the sum of longitudinal and transverse components. We also make use of the Fourier transforms defined in Eqs. (17a) and (17b) to move the temporal operators to the frequency domain, giving

$$(\partial_z^2 + \nabla_{\perp}^2) \hat{E} + k^2(\omega) \hat{E} = -\frac{\omega^2}{\epsilon_0 c^2} \hat{N}_p, \quad (18)$$

where

$$k(\omega) \equiv \frac{\omega \sqrt{\epsilon(\omega)}}{c}, \quad (19)$$

and

$$\hat{N}_p = \mathcal{F}\{N_p E\}. \quad (20)$$

Our interest in this paper is the propagation of optical *pulses*. As such we introduce the concept of an electromagnetic pulse per Born and Wolf[6].

$$E(\mathbf{r}, t) = \Re \left\{ \int_{-\infty}^{\infty} a_{\omega}(\mathbf{r}) e^{-i[\omega t - g_{\omega}(\mathbf{r})]} d\omega \right\} \quad (21)$$

where  $\Re\{\}$  is the *real part of operator*, describes an optical *pulse* or *wave group* if the phase function  $g_{\omega}(\mathbf{r})$  can be meaningfully approximated by a linear function of frequency  $\omega$  over the range where amplitude  $a_{\omega}(\mathbf{r})$  differs appreciably from zero. When this is the case, the concept of a *pulse* can be used to model the propagation of an electromagnetic disturbance which retains some sense of limited support (the region over which it is non-zero) in space and time as it moves. This may be seen via the *Fourier Shift Theorem* (see Bracewell [10]) which states that translation of a function in the spatial or temporal domain corresponds to a linear phase shift in its Fourier transformed distribution [10]. Eq. (21) describes the instantaneous electric field as a function of space and time. In what follows, we use the typical convention of discussing the *complex field* (without taking the real part of the right hand side in Eq. (21)). Ultimately we develop propagation equations in terms of a *complex pulse envelope* in order to make the numerical modeling problem feasible.

As we observe the optical disturbance propagate in a dispersive material the pulse description given in Eq. (21) gives rise to two concepts of velocity. Phase velocity describes the motion of the *surfaces of constant phase* in the wave group, while group velocity describes the rate at which *surfaces of constant amplitude* travel. We may also view the group velocity as a description of how fast the wave envelope travels. In a non-dispersive medium the group and phase velocities are the same. *Phase velocity* and *group velocity* are given respectively as

$$v_p = \frac{\omega}{k} \quad \text{and} \quad v_g = \frac{d\omega}{dk} = \left( \frac{dk}{d\omega} \right)^{-1} \equiv \frac{1}{k'}, \quad (22)$$

where the final definition may be understood with the help of Eq. (41). We later transform our propagation physics into a coordinate system that moves at the group velocity so that our numerical grid moves along with the pulse (eliminating the need for a grid covering all of the space between source and destination).

Having established that the pulse must have effectively finite support in the temporal frequency domain we may define a *center frequency*. The principal purpose of the center frequency definition is to allow us

to later “factor out” or “eliminate” a fast phase component of our description thus facilitating a discrete numerical model with lower sampling requirements. We take Brabec’s[9] definition given as

$$\omega_o \equiv \frac{\int_o^\infty d\omega \omega |\hat{E}(\omega)|^2}{\int_o^\infty d\omega |\hat{E}(\omega)|^2}, \quad (23)$$

Naturally, during nonlinear propagation the center frequency per Eq. (23) will change as the pulse spectrum is altered. Hence,  $\omega_o$  really represents the center frequency of the pulse *before the onset of nonlinear propagation*.

We now transform to a coordinate system traveling at the group velocity of the pulse  $(t, z) \rightarrow (\tau, \zeta)$ , where

$$\tau = t - k'z, \quad \zeta = z. \quad (24)$$

From the chain rule we obtain

$$\partial_t = \partial_\tau, \quad \partial_z = \partial_\zeta - k'\partial_\tau \quad (25)$$

The temporal frequency domain representations of the operators in Eq. (25) are

$$\partial_t = -i\omega, \quad \partial_z = \partial_\zeta + ik'\omega. \quad (26)$$

We now express Eq. (18) in the pulse reference frame via operator substitution as

$$[(\partial_\zeta + ik'\omega)^2 + \nabla_\perp^2]\hat{E} + k^2(\omega)\hat{E} = -\frac{\omega^2}{\epsilon_o c^2}\hat{N}_p, \quad (27)$$

where  $\hat{E} \equiv \hat{E}(\mathbf{r}, \tau, \zeta)$ . Eq. (27) is a carrier-resolved description in the pulse reference frame expressed in the temporal frequency domain. We refer to the center frequency described in Eq. (23) as the carrier. In order to facilitate subsequent development we expand one of the operators in Eq. (27) to obtain

$$\partial_\zeta^2 \hat{E} + 2ik'\omega \partial_\zeta \hat{E} = -\nabla_\perp^2 \hat{E} - [k^2(\omega) - (k'\omega)^2]\hat{E} - \frac{\omega^2}{\epsilon_o c^2}\hat{N}_p. \quad (28)$$

Ultimately our model will be written in terms of envelope variables in order to create a computationally viable framework. We now introduce the envelope models. For the laboratory fixed frame we write

$$E(\mathbf{r}, t, z) = \Re\{\mathcal{E}(\mathbf{r}, t, z) \exp(-i\omega_o t + ikz)\}. \quad (29)$$

From Eq. (29) via Eqs. (24) we have

$$E(\mathbf{r}, \tau, \zeta) = \Re\{\mathcal{E}(\mathbf{r}, \tau, \zeta) \exp(-i\omega_o \tau + i(k - k'\omega_o)\zeta)\}, \quad (30)$$

for the moving pulse frame. In Eqs. (29) and (30) the quantity  $E$  represents the instantaneous scalar electric field amplitude while  $\mathcal{E}$  is the *envelope* of the electric field.

Envelope models are useful when two distinct temporal scales exist in a propagation problem. They allow us to separate a fast scale (oscillation of the instantaneous electric field amplitude) from a slow scale (overall shape of the optical pulse). The envelope is complex, allowing phase variation (deviation from the fast phase) over the duration of the pulse to be described. The complete electric field can be reconstructed from this envelope easily as the real part of the product of the envelope and the fast phase component. In order to arrive at a propagation relationship expressed in terms of only envelope variables we will need to substitute each envelope term into Eq. (28) accounting for the effect of the first and second order derivatives with respect to  $\zeta$  on the envelope expression. Using Eq. (30) we obtain the following general relationship for the  $n$ th derivative of the envelope with respect to  $\zeta$ ,

$$\partial_\zeta^n E = \exp(-i\omega_o \tau + i(k - k'\omega_o)\zeta) [\partial_\zeta + i(k - k'\omega_o)]^n \mathcal{E}, \quad (31)$$

with Fourier transform in the temporal variable  $\tau$

$$\partial_\zeta^n \hat{E} = \exp(i(k - k'\omega_o)\zeta) [\partial_\zeta + i(k - k'\omega_o)]^n \hat{\mathcal{E}}, \quad (32)$$

where  $\hat{\mathcal{E}}$  is a function of  $\omega - \omega_o$  (see Bracewell [10]). Assuming a similar envelope model for the nonlinear polarization term (see [12], page 30), we may use Eq. (32) and Eq. (28) to obtain (after canceling the ubiquitous complex exponential term)

$$\partial_\zeta^2 \hat{\mathcal{E}} + 2i\kappa(\omega)\partial_\zeta \hat{\mathcal{E}} = -\nabla_\perp^2 \hat{\mathcal{E}} - (k^2(\omega) - \kappa^2(\omega))\hat{\mathcal{E}} - \frac{\omega^2}{c^2\epsilon_o}\hat{\mathcal{N}}_p \quad (33)$$

where  $\kappa(\omega) = k + k'(\omega - \omega_o)$ , and

$$\hat{\mathcal{N}}_p = \mathcal{F}\{\mathcal{N}_p\mathcal{E}\} \quad (34)$$

Note that  $\kappa(\omega)$  is really the sum of the first two terms in the Taylor series expansion of  $k(\omega)$ . In writing Eq. (33) using variable envelopes we have presumed that it is possible to write the nonlinear operators in envelope form. In section 2.2 we show that this is possible.

We make an essential approximation that produces an initial value problem from the second order partial differential Eq. (33) by requiring

$$|\partial_\zeta^2 \hat{\mathcal{E}}| \ll 2\kappa(\omega)|\partial_\zeta \hat{\mathcal{E}}|. \quad (35)$$

Eq. (35) indicates that the magnitude of the *change* in  $\partial_\zeta \hat{\mathcal{E}}$  is *small* when compared with the actual magnitude of  $\partial_\zeta \hat{\mathcal{E}}$  scaled by  $\kappa(\omega)$  (which is approximately inversely proportional to the spatial wavelength). In other words, the magnitude and phase of the pulse envelope *evolve* slowly with respect to the motion of the pulse in the  $\zeta$ -direction. In order to maintain slow evolution of the pulse as we move in the principle direction of propagation, we cannot allow angular spectrum components with large angles. This implies a paraxial model (similar to the assumption used to obtain Eq. (14)).

Using the approximation in Eq. (35) to simplify Eq. (33) yields

$$\partial_\zeta \hat{\mathcal{E}} = \frac{i}{2\kappa(\omega)}\nabla_\perp^2 \hat{\mathcal{E}} + i\frac{k^2(\omega) - \kappa^2(\omega)}{2\kappa(\omega)}\hat{\mathcal{E}} + i\frac{\omega^2}{2\kappa(\omega)c^2\epsilon_o}\hat{\mathcal{N}}_p. \quad (36)$$

Eq. (36) is *one-way*, meaning that it presumes reflected waves are insignificant (see Feit and Fleck [17]). Also, we have placed restrictions only on the evolution of the pulse in the  $\zeta$ -direction, meaning that steep changes in the envelope along the temporal axis are allowed.

The frequency content of the pulse envelope is naturally much lower than that of the pulse itself (meaning  $\Delta\omega \ll \omega_o$ ). Specifically, factoring out the carrier has left a pulse envelope description centered about the carrier frequency (the carrier frequency is at the origin of a spectral plot of the envelope). The envelope frequencies are really differences from the carrier and the frequency axis relationship between the envelope and the pulse is  $\Omega = \omega - \omega_o$ .

The first term on the right-hand side of Eq. (36) operates on transverse coordinates only and represents diffraction. The last term represents the nonlinear material response. The center term is our dispersion model. It represents the *linear* material response. We know that  $\kappa(\omega)$  is the sum of the first two terms in the Taylor series expansion of  $k(\omega)$ . The difference between  $\kappa(\omega)$  and  $k(\omega)$  is comprised of higher order (second and above) Taylor series terms. Thus we know that the expression

$$\frac{k^2(\omega) - \kappa^2(\omega)}{2\kappa(\omega)}, \quad (37)$$

describes the dispersion of the envelope under our model. The ratio of angular frequency to wavenumber gives the corresponding phase velocity. We know that in our moving coordinate system the phase velocities of each frequency must be adjusted by the velocity of the numerical grid (which is by design the group velocity of the pulse). Frequencies traveling at the group velocity should have phase velocities of zero. Those traveling faster should have positive phase velocities. Those traveling slower must have negative phase velocities so that they can move backward on our computational grid. With a little algebra we may approximate the dispersion relationship in Eq. (37) as

$$\begin{aligned} \frac{k^2(\omega) - \kappa^2(\omega)}{2\kappa(\omega)} &= [k(\omega) - \kappa(\omega)] \left( 1 + \frac{k(\omega) - \kappa(\omega)}{2\kappa(\omega)} \right) \\ &\approx k(\omega) - \kappa(\omega), \end{aligned} \quad (38)$$

where the neglected terms are necessarily fourth order in  $\Omega$ . Using Eq. (38) in Eq. (36) gives

$$\partial_\zeta \hat{\mathcal{E}} = \frac{i}{2\kappa(\omega)} \nabla_\perp^2 \hat{\mathcal{E}} + i(k(\omega) - \kappa(\omega)) \hat{\mathcal{E}} + i \frac{\omega^2}{2\kappa(\omega)c^2\epsilon_o} \hat{\mathcal{N}}_p. \quad (39)$$

The dispersion relationship in Eqs. (38) and (39) may also be expressed as a Taylor series expansion

$$D(\Omega) \equiv k(\omega) - \kappa(\omega) = \sum_{m=2}^{\infty} \frac{k^{(m)}}{m!} \Omega^m, \quad (40)$$

where the wavenumber

$$k^{(m)} = \left. \frac{\partial^m k(\omega)}{\partial \omega^m} \right|_{\omega=\omega_o}. \quad (41)$$

The sum in Eq. (40) begins at  $m = 2$  because  $\kappa(\omega)$  subtracts off the first two terms of the series. Often the first three dispersion coefficients defined by Eq. (41) are written as  $k'$ ,  $k''$ , and  $k'''$  respectively and we adhere to that convention in our development.

We now transform Eq. (39) back into the time domain. We can express everything in terms of the slow frequency variable  $\Omega = \omega - \omega_o$ . Remember that

$$\partial_\tau \rightarrow -i\Omega \quad \text{or} \quad i\partial_\tau \rightarrow \Omega \quad (42)$$

so that

$$\omega = \omega_o + \Omega \rightarrow \omega_o \left( 1 + \frac{i}{\omega_o} \partial_\tau \right). \quad (43)$$

Also,

$$\begin{aligned} \kappa(\omega) &= k + k'\Omega \rightarrow k \left( 1 + i \frac{k'}{k} \partial_\tau \right) \\ &\approx k \left( 1 + \frac{i}{\omega_o} \partial_\tau \right), \end{aligned} \quad (44)$$

where the final approximation assumes the group velocity is nearly equal to the phase velocity. If we identify the shock operators

$$T \equiv \left( 1 + \frac{i}{\omega_o} \partial_\tau \right) \quad \text{and} \quad S \equiv \left( 1 + i \frac{k'}{k} \partial_\tau \right), \quad (45)$$

we then have

$$\partial_\zeta \mathcal{E} = \frac{i}{2k} S^{-1} \nabla_\perp^2 \mathcal{E} + iD(i\partial_\tau) \mathcal{E} + \frac{i\omega_o}{2n_o c \epsilon_o} S^{-1} T^2 \mathcal{N}_p \mathcal{E}, \quad (46)$$

where we have used  $ck = n_o \omega_o$ . Using Eq. (44) we simplify the propagation description to

$$\partial_\zeta \mathcal{E} \approx \frac{i}{2k} T^{-1} \nabla_\perp^2 \mathcal{E} + iD(i\partial_\tau) \mathcal{E} + \frac{i\omega_o}{2n_o c \epsilon_o} T \mathcal{N}_p \mathcal{E}. \quad (47)$$

In order to obtain nonlinear terms with forms familiar from the literature it is convenient to define

$$\mathcal{N} \equiv \frac{ik_o}{2n_o \epsilon_o} \mathcal{N}_p \quad (48)$$

and write

$$\partial_\zeta \mathcal{E} \approx \frac{i}{2k} T^{-1} \nabla_\perp^2 \mathcal{E} + iD(i\partial_\tau) \mathcal{E} + T \mathcal{N} \mathcal{E} \quad (49)$$

Often only the first two terms ( $m = 2, 3$ ) in the summation  $D(i\partial_\tau)$  are retained. These are called *group velocity dispersion* and *third order dispersion* respectively.

## 2.2 Nonlinear Material Model

In this section we outline the physics behind the nonlinear portion of the mathematical model. Section 2.2.1 discusses the nearly instantaneous refractive index perturbation introduced into the medium by the presence of the strong electric field associated with a high intensity optical disturbance (referred to as the Kerr effect). We derive an expression for the nonlinear refractive index (the constant of proportionality relating the cube of the field value to the nonlinear polarization of the medium). This nonlinear polarization directly affects the propagation of the optical pulse via wave front delay caused by the increase in refractive index or decrease in wavespeed in high-intensity regions of the pulse and is primarily responsible for self-focusing. Section 2.2.2 describes stimulated Raman scattering. This effect is also third order in field value. It is a delayed rather than an instantaneous effect and is represented by a convolution operator.

### 2.2.1 Instantaneous Kerr Effect

The Kerr effect represents nonlinear electronic polarization that takes place nearly instantaneously. Centrosymmetry (crystallographic or molecular) refers to a distribution containing an inversion center as one of its symmetry elements. For every point  $(x, y, z)$  in the distribution (unit cell for solids and molecule for liquids and gasses) there is an indistinguishable location  $(-x, -y, -z)$ . The absence of this type of symmetry is required in order to display properties such as the piezoelectric effect, the Pockels effect, optical rectification, etc. Centrosymmetric materials have an electronic polarization response that can be described via a Taylor series expansion in odd powers.

$$\mathbf{P} = \epsilon_o \chi^{(1)} \mathbf{E} + \epsilon_o \chi^{(3)} (\mathbf{E} \cdot \mathbf{E}) \mathbf{E} + \dots \quad (50)$$

The first order term gives rise to the concept of refractive index. Its frequency dependence introduces the concept of dispersion and results in the convolution integral in Eq. (11) and the Taylor expansion of time-derivatives in Eqs. (46) and (49). For the nonlinear polarization operator expression in a centrosymmetric material we have

$$\mathbf{N}_p \mathbf{E} = \epsilon_o \chi^{(3)} (\mathbf{E} \cdot \mathbf{E}) \mathbf{E}. \quad (51)$$

Using the envelope model (either lab or pulse frame written as the average of complex conjugates) we may write

$$E(\mathbf{r}, t, z) = \frac{1}{2} (\mathcal{E}(\mathbf{r}, t, z) \exp(-i\omega_o t + ikz) + \text{c.c.}), \quad (52)$$

where c.c. refers to the complex conjugate of the first term. Eqs. (52) and (51) give

$$\begin{aligned} E^3(\mathbf{r}, t, z) &= \frac{1}{8} (\mathcal{E}^3(\mathbf{r}, t, z) \exp(-i3\omega_o t + i3kz) \\ &\quad + 3|\mathcal{E}|^2 \mathcal{E} \exp(-i\omega_o t + ikz) + \text{c.c.}). \end{aligned} \quad (53)$$

Each term of an envelope model is written as the product of a slowly varying term and a fast phase term (complex exponential) with common frequency  $\omega_o$  (see Eqs (29) and (30)). Eq. (53) has a complex exponential term with frequency  $3\omega_o$  (corresponding to the physical phenomenon of third harmonic generation). In order to obtain a complete envelope model devoid of any fast phase components we must neglect the third harmonic term. We now have

$$E^3(\mathbf{r}, t, z) = \frac{3}{8} |\mathcal{E}|^2 \mathcal{E} \exp(-i\omega_o t + ikz) + \text{c.c.} \quad (54)$$

This allows us to write the polarization as a simple envelope function, giving

$$P(\mathbf{r}, t, z) = \frac{1}{2} (\mathcal{P}(\mathbf{r}, t, z) \exp(-i\omega_o t + ikz) + \text{c.c.}), \quad (55)$$

from which we infer

$$\mathcal{P}(\mathbf{r}, t, z) = \frac{3}{4} \epsilon_o \chi^{(3)} |\mathcal{E}|^2 \mathcal{E}. \quad (56)$$

The above development uses electric field units of (V/m) rather than the  $\sqrt{W}/m$ . The definition of the nonlinear refractive index in terms of basic material parameters varies widely in the literature <sup>1</sup>. A natural definition from the expression in Eq. (56) might be simply  $3\chi^{(3)}/4$ . However, it is common in the literature to write the total refractive index as the sum of the linear index and the product of the nonlinear index and intensity (see for example Diels [15]). We therefore explicitly define

$$n_2 \equiv \frac{3\chi^{(3)}}{4\epsilon_o c n_o^2}, \quad (57)$$

with units of  $m^2/W$ . In doing so we use the Poynting vector relationship between electric field and intensity [26]. In SI units for instantaneous fields and plane waves the intensity is equal to the square of electric field times the reciprocal of the free-space impedance. If the fields are time-harmonic we may average over a single cycle of the variation and now the intensity is one-half the magnitude of electric field times the reciprocal of the free-space impedance. For envelope models the time-harmonic relationship cannot hold perfectly (the envelope is a function of time and must remain under the integral during our average over a carrier cycle). However, the approximation

$$\mathcal{I} \approx \frac{c n_o \epsilon_o}{2} |\mathcal{E}|^2 \quad (58)$$

is good down to very short pulses. We can now write

$$\mathcal{P}(\mathbf{r}, t, z) = 2\epsilon_o n_o n_2 \mathcal{I} \mathcal{E} \quad (59)$$

Using the scaling in Eq. (48) the operator expression becomes

$$\mathcal{N}_k \mathcal{E} \equiv i k_o n_2 \mathcal{I} \mathcal{E}. \quad (60)$$

It should be evident at this point in our development that we have assumed a centrosymmetric material and neglected third-harmonic generation.

### 2.2.2 Raman Scattering

Stimulated molecular Raman scattering is inelastic. It results from interaction with rotational and vibrational modes of the molecule and (like the Kerr effect) is quadratic in the field value. Even though the physical origins are different, we often combine the Kerr effect and stimulated Raman scattering because their dependence on field strength is identical. The contribution of stimulated Raman scattering to the polarization of the material may be modeled as a driven, damped oscillator via a differential equation in polarizability (see Couairon [13])

$$\partial_t^2 \mathcal{Q}_R + 2\gamma \partial_t \mathcal{Q}_R + (\omega_R^2 + \gamma^2) \mathcal{Q}_R = (\omega_R^2 + \gamma^2) \mathcal{I}, \quad (61)$$

where  $\mathcal{Q}_R$  is a kind of generalized material polarization response with units of intensity,  $\omega_R$  is the Raman frequency, and  $\gamma$  is an empirically determined damping rate. The driving force is quadratic in the external field. With boundary conditions that require the polarization to go to zero at positive and negative infinity, we have a solution of

$$\mathcal{Q}_R(\mathbf{r}, t, z) = \int_{-\infty}^t R(\tau) \mathcal{I}(\mathbf{r}, t - \tau, z) d\tau, \quad (62)$$

with

$$R(t) = \frac{\gamma^2 + \omega_R^2}{\omega_R} \exp(-\gamma t) \sin(\omega_R t). \quad (63)$$

We may now sum the total Kerr and Raman scattering responses to obtain

$$\mathcal{P}_{kr} = 2\epsilon_o n_o n_2 [(1 - \alpha) \mathcal{I} + \alpha \mathcal{Q}_R] \mathcal{E}, \quad (64)$$

---

<sup>1</sup>See for example Agrawal [3], page 33, Eq. 2.3.13 or Couairon [1], page 194 in the paragraph just after Eq. 2. Also see Sutherland [35], page 345, Table 4.

where  $\mathcal{P}_{kr}$ ,  $\mathcal{E}$ ,  $\mathcal{I}$ , and  $\mathcal{Q}_R$  are understood to be functions of  $(\mathbf{r}, t, z)$  and the fractional portion of the combined effect due to Raman scattering is given as  $\alpha$ . Using the scaling in Eq. (48) we write the operator expression as

$$\mathcal{N}_{kr}\mathcal{E} \equiv ik_on_2[(1-\alpha)\mathcal{I} + \alpha\mathcal{Q}_R]\mathcal{E}. \quad (65)$$

### 2.2.3 Ionization

When field values are high enough the effects of ionization must appear in our model in several places. We need an evolution equation that describes how changing field values, recombination, multiphoton ionization (MPI), etc., drive the free-electron density  $\rho$ . As the high field values in the pulse ionize the medium it loses energy. We need to express absorption in the propagator representing the removal of energy from the pulse due to ionization. The refractive index generally decreases with increasing free-electron density. Refractive index variation with change in the free-electron density must be accounted for in the principal propagation equation. Linear and nonlinear shock terms are included in this development. A comparison of Eqs. (76) and (89) demonstrates that the inclusion of ionization impedance effects alters the shock operator in expressions for plasma defocusing. This renders the final simulation equation different not only from some treatments without shock (see [29, 31, 32]) but also from some which do include both shock and ionization [14, 13, 27].

**Free-electron Density** We first describe the evolution of the free-electron number density  $\rho$  via

$$\frac{d\rho}{dt} = \frac{\sigma}{n_o^2 U_i} \rho \mathcal{I} + \mathcal{I}^K \sigma_k (\rho_{nt} - \rho) - \alpha_r \rho^2, \quad (66)$$

where

- $\mathcal{I}$  = Electric field intensity
- $n_o$  = Background refractive index
- $K$  = Photons required to free single electron
- $U_i$  = Energy required to free single electron
- $\rho_{nt}$  = Neutral atom number density
- $\alpha_r$  = Electron-positive ion recombination loss
- $\sigma_k$  = Ionization cross-section.

The first term in Eq. (66) is the avalanche or cascade ionization. The second is multiphoton ionization (MPI) tempered with a saturation term. We have assumed that the ionization cross-section is constant and takes on a value characteristic of MPI. Physically, it actually exhibits a dependence on field intensity [34]. At high field strengths its value must be chosen to represent the more prevalent tunneling ionization. The final term represents the production of neutral atoms through electron-ion recombination. It is possible under the Drude model to obtain an approximate expression for  $\sigma$ , the avalanche ionization rate coefficient, in terms of the electron relaxation time and the center frequency of the pulse. However, this relationship does not always hold so we choose to allow these as separate parameters.

In cases where the neutral atom density is not known precisely and it is possible to neglect saturation effects, we may use

$$\frac{d\rho}{dt} = \frac{\sigma}{n_o^2 U_i} \rho \mathcal{I} + \frac{\beta_K}{K \hbar \omega_o} \mathcal{I}^K - \alpha_r \rho^2, \quad \rho \ll \rho_{nt} \quad (67)$$

per Berge [28]. The parameter  $\beta_K$  is typically specified directly, but is related to  $\sigma_k$  and the unknown neutral atom number density via

$$\beta_K = \sigma_k \rho_{nt} K \hbar \omega_o. \quad (68)$$

**Plasma Defocusing** As part of developing the nonlinear material model we need an expression for the time derivative of electric current (see Eqs. (15) and (16)). This term ultimately becomes what is known as the



plasma defocusing term. Current  $\mathbf{J}$  is the flow of charge through a surface per unit time. The total charge per unit volume is the product of electron number density (electrons per unit volume) and the charge per electron. The quantity of charge flowing through a unit area is the product of total charge density and the rate of flow or velocity giving

$$\mathbf{J} = -e\rho\mathbf{v}, \quad (69)$$

where  $e$  is the charge on an electron,  $\rho$  is the total free-electron number density and  $\mathbf{v}$  is the velocity of the flow. Differentiating both sides of Eq. (69) gives

$$\frac{\partial\mathbf{J}}{\partial t} = -e\rho\frac{\partial\mathbf{v}}{\partial t}. \quad (70)$$

The value of the electric field is defined as [22]

$$\mathbf{E} = \frac{\mathbf{F}}{q}, \quad (71)$$

where  $\mathbf{F}$  is the force in Newtons and  $q$  is a quantity of charge in Coulombs. Using Eqs. (70) and (71) Newton's second law gives

$$\frac{d\mathbf{v}}{dt} = \frac{\mathbf{F}}{m} = -\frac{e}{m_e}\mathbf{E}. \quad (72)$$

Using Eq. (72) in Eq. (70) gives

$$\frac{\partial\mathbf{J}}{\partial t} = \frac{e^2}{m_e}\rho\mathbf{E} \quad (73)$$

We must express the effect of the ionization current in Eq. (73) as an equivalent polarization for use in our nonlinear model. We may obtain the necessary expression for the *integral* of the current density (see Eq. (16)) most easily in the frequency domain. Using the frequency domain envelope representation we compute the Fourier transform of the integral of the current density as

$$\hat{\mathcal{P}}_{\text{pls}} = \frac{1}{(-i\omega)^2}\mathcal{F}\left\{\frac{e^2}{m_e}\rho\mathcal{E}\right\}, \quad (74)$$

where division by  $-i\omega$  represents the effect of integration. Moving back into the slow-time (or temporal envelope) domain we have the following expression for the plasma current equivalent polarization

$$\mathcal{P}_{\text{pls}} = -\frac{1}{(\omega_o T)^2}\frac{e^2}{m_e}\rho\mathcal{E}. \quad (75)$$

Expressing Eq. (75) in a form convenient for use in our nonlinear propagation equation we have

$$T^{-1}\mathcal{N}_{\text{pls}}\mathcal{E} = -\frac{ik_o}{2n_o\rho_c}T^{-2}(\rho\mathcal{E}), \quad (76)$$

where we have used the definition of the critical plasma density

$$\rho_c \equiv \epsilon_o m_e \omega_o^2 / e^2, \quad (77)$$

above which the plasma becomes opaque to waves with frequencies below  $\omega_o$ . Note the double shock operator present in Eq. (76).

The relationship in Eq. (76) assumes electrons move in the presence of an applied electric field but does not take into account collisions between the electrons and the positive ions in our plasma. Drude proposed a model for electrons in metals that assumes a sea of other particles that function as hard, essentially stationary scattering centers. This model is certainly applicable for ionization in gases. The larger, positive ions do not respond very much at all to an oscillating electric field. For this reason the Drude model is sometimes used to describe this portion of the current. In particular it is needed in order to move from our instantaneous description in Eq. (73) to our envelope model.

We now develop an impedance relationship for the plasma that relates current and electric field. The result will be loss and delay terms that affect the propagation of the pulse. These naturally depend on the strength of the electric field through the generation of the plasma electron density. We develop an equation for the evolution of average electron momentum  $\mathbf{p}(t)$ . During a *short* time interval  $dt$  the probability that a given electron will experience a collision is approximately  $dt/\tau_c$  where  $\tau_c$  is known as the relaxation time (considered to be independent of the position or current velocity of the electron). This means that the change in momentum may be written

$$\mathbf{p}(t + dt) - \mathbf{p}(t) = -\frac{dt}{\tau_c} \mathbf{p} \quad (78)$$

Eq. (78) is a simple relaxation equation. In the absence of any perturbing forces (such as electric fields), we may write the evolution of momentum as the product of an initial value and a simple decaying exponential with time constant  $\tau_c$  (hence the name *relaxation time*). If we have an electric field present then we simply add a term representing the change in momentum  $\mathbf{F}dt$  to arrive at

$$\begin{aligned} \mathbf{p}(t + dt) - \mathbf{p}(t) &= -\frac{dt}{\tau_c} \mathbf{p}(t) + \left(1 - \frac{dt}{\tau_c}\right) \mathbf{F}dt \\ &\approx -\frac{dt}{\tau_c} \mathbf{p}(t) + \mathbf{F}dt, \end{aligned} \quad (79)$$

where we have neglected the second order term in  $dt$ . Finally we may divide both sides of Eq. (79) by  $dt$ , use the fact that  $\mathbf{F} = -e\mathbf{E}$  and take a limit as the time interval approaches zero to yield

$$\frac{d\mathbf{p}(t)}{dt} = -\frac{\mathbf{p}(t)}{\tau_c} - e\mathbf{E}. \quad (80)$$

Next we assume a time-harmonic solution to Eq. (80). Our pulses are not time-harmonic, they are envelope based and so the relationship we obtain between the electric field and momentum (and subsequently current) will be an approximate one.

$$\mathbf{E}(t) = \frac{1}{2} [\mathbf{E}(\omega) \exp(-i\omega t) + \mathbf{E}^*(\omega) \exp(i\omega t)] \quad (81)$$

$$\mathbf{p}(t) = \frac{1}{2} [\mathbf{p}(\omega) \exp(-i\omega t) + \mathbf{p}^*(\omega) \exp(i\omega t)] \quad (82)$$

Substituting Eqs. (81) and (82) into Eq. (80) and solving for momentum we obtain

$$\mathbf{p}(\omega) = \frac{-e\tau_c}{1 - i\omega\tau_c} \mathbf{E}(\omega) \quad (83)$$

The linearity of Eq. (80) gives a separate equation for the conjugate of momentum which we don't really need. Eq. (69) tells us that we can relate momentum to current and write

$$\begin{aligned} \mathbf{J}(\omega) &= \left(-\frac{e\rho}{m_e}\right) \mathbf{p}(\omega) = \left(-\frac{e\rho}{m_e}\right) \frac{-e\tau_c}{1 - i\omega\tau_c} \mathbf{E}(\omega) \\ &\approx \left(\frac{e^2\rho\tau_c}{m_e}\right) \frac{1 + i\omega\tau_c}{1 + \omega^2\tau_c^2} \mathbf{E}(\omega) \end{aligned} \quad (84)$$

Eq. (84) represents a full spectral response by the current to the electric field. The real part accounts for plasma absorption and the imaginary part for plasma defocusing. These effects vary with frequency and in order to model the dispersive nature of the plasma impedance we would need to solve Eq. (84) at each step and update each frequency component of the pulse accordingly. If we accept the impedance relationship at the center frequency of the pulse as holding over the entire pulse bandwidth we may simplify Eq. (84) to

$$\mathbf{J}(\omega) \approx \left(\frac{e^2\rho\tau_c}{m_e}\right) \frac{1 + i\omega_o\tau_c}{1 + \omega_o^2\tau_c^2} \mathbf{E}(\omega), \quad (85)$$

by replacing the general frequency variable  $\omega$  with the constant  $\omega_o$ . We may simplify the model somewhat by combining the nonlinear polarization and the plasma current into a single term (see Eqs. (16) and (18)) and consider the relationship only at the center frequency of the pulse. We then require the integral of current density

$$\mathbf{P}_{\text{pls}}(\omega) = \frac{\mathbf{J}(\omega)}{-i\omega} \approx -\frac{1}{i\omega} \left( \frac{e^2 \rho \tau_c}{m_e} \right) \frac{1 + i\omega_o \tau_c}{1 + \omega_o^2 \tau_c^2} \mathbf{E}(\omega). \quad (86)$$

In this case we neglect the dispersive or frequency dependent nature of the relationship between the electric field and the integral of the current and use only the complex impedance at the center frequency of the pulse. Using the envelope representation we write the term we add to the nonlinear polarization as

$$\hat{\mathcal{P}}_{\text{pls}} = -\frac{1}{i\omega} \left( \frac{e^2 \rho \tau_c}{m_e} \right) \frac{1 + i\omega_o \tau_c}{1 + \omega_o^2 \tau_c^2} \hat{\mathcal{E}} \quad (87)$$

Now we move Eq. (87) into the slow-time domain via Eq. (43)

$$\mathcal{P}_{\text{pls}} = -\frac{1}{i\omega_o T} \left( \frac{e^2 \rho \tau_c}{m_e} \right) \frac{1 + i\omega_o \tau_c}{1 + \omega_o^2 \tau_c^2} \mathcal{E} \quad (88)$$

as was done for the propagation Eq. (49) in section 2.1.

Using the scale factor defined in Eq. (48) we write the operator expression as

$$T^{-1} \mathcal{N}_{\text{pls}} \mathcal{E} \equiv -\frac{\sigma}{2} (1 + i\omega_o \tau_c) T^{-1} (\rho \mathcal{E}) \quad (89)$$

where

$$\sigma \equiv \frac{k_o}{n_o \rho_c} \frac{\omega_o \tau_c}{1 + \omega_o^2 \tau_c^2}, \quad (90)$$

is the cross-section for inverse Bremsstrahlung (the process by which an electron absorbs energy from the optical pulse during a collision). Note that the power of the shock operator in Eq. (89) differs from that in Eq. (76).

**MPI Energy Loss** We need a term in our propagation equation to represent the reduction in optical pulse energy required to ionize the medium (known as multiphoton absorption or MPA). We do this by constructing an artificial current with the value necessary to dissipate the needed energy. This current is the coupling between the propagation or field Eq. (49) and the ionization physics model in Eq. (66). In order to obtain the expression for the coupling we seek the value of a current which will dissipate an amount of power per unit volume equal to that required to support the ionization. Remember that work is defined as

$$W = \int \mathbf{F} \cdot d\mathbf{x} = \int \mathbf{F} \cdot \mathbf{v} dt, \quad (91)$$

and thus

$$P = \frac{dW}{dt} = \mathbf{F} \cdot \mathbf{v}. \quad (92)$$

By definition the force on a charge in an electric field is the product of the charge and electric field values. Hence, force per unit volume is the product of charge density and electric field value. Finally, the power per unit volume  $S$  (via Eq. (92)) is

$$S = -e\rho \mathbf{v} \cdot \mathbf{E} = \mathbf{J} \cdot \mathbf{E}, \quad (93)$$

where the last equality results from the fact that the product of a density and a velocity is a flux. This may be seen by considering a thin slice (taken normal to the velocity vector) through the density, of thickness  $dx$ . If this slice moves through its front face in time  $dt$  then it is traveling with velocity  $dx/dt = v$ . The product of the density and velocity is  $\rho dx/dt$  which in our case gives how many Coulombs of charge pass through the front face of our slice each second. If we postulate a current of the right value flowing in our electric field then we will naturally have the correct power dissipation. Let us assume that the ionization rate is directly

proportional to the number of available neutral atoms  $\rho_{\text{nt}}$  and that we may neglect saturation effects. We write

$$\frac{d\rho}{dt} = W_r \rho_{\text{nt}}, \quad (94)$$

where  $W_r$  is the photoionization rate. This gives the number of new free electrons we should expect per unit volume per second. For each of these electrons  $K$  photons were absorbed, each having an energy of  $\hbar\omega$ . Hence the total energy needed *per unit volume per second* is given by

$$S = W_r \rho_{\text{nt}} K \hbar \omega_o = \mathbf{J} \cdot \mathbf{E}. \quad (95)$$

This implies that the current density must be

$$\mathbf{J} = \frac{W_r \rho_{\text{nt}} K \hbar \omega_o}{|\mathbf{E}|^2} \mathbf{E}, \quad (96)$$

where the normalization by the square of the electric field is simply to cancel the similar term in the numerator of the dot product of current density and electric field. We have expressed everything up to now in terms of the instantaneous fields. Using an approximation similar to that in Eq. (58) we note that

$$\mathcal{S} \approx \frac{1}{2} \mathcal{J} \cdot \mathcal{E} \quad (97)$$

and construct an expression similar to that in Eq. (96) for our developed envelope equation

$$\mathcal{J} = 2 \frac{W_r \rho_{\text{nt}} K \hbar \omega_o}{|\mathcal{E}|^2} \mathcal{E}, \quad (98)$$

where the factor of two compensates for the one-half in Eq. (97).

We need a model for the photoionization rate  $W_r$ . For a  $K$ -photon ionization process a commonly used model is

$$W_r = \sigma_k \mathcal{I}^K, \quad (99)$$

giving

$$\mathcal{J} = 2 \frac{\sigma_k \rho_{\text{nt}} K \hbar \omega_o \mathcal{I}^K}{|\mathcal{E}|^2} \mathcal{E} = 2 \frac{\beta_K \mathcal{I}^K}{|\mathcal{E}|^2} \mathcal{E} \quad (100)$$

where we have used Eq. (68).

Just as we did in section 2.2.3 we seek a term that can be added to the polarization (even though we are modeling the loss with a current). Again, we need the *integral* of the current and so we use the same technique as we did in obtaining Eqs. (75) and (88). Expressing the relationship between current and electric field in the frequency domain, dividing by  $(-i\omega)$  to obtain the integral of current and transforming back into the slow-time temporal envelope domain we have

$$\mathcal{P}_{\text{MPA}}(\mathbf{r}, t, z) = T^{-1} 2 \frac{\beta_K \mathcal{I}^K}{-i\omega_o |\mathcal{E}|^2} \mathcal{E}, \quad (101)$$

for the equivalent material polarization representing MPA. Using Eqs. (58), (48) and (101) we write

$$T^{-1} \mathcal{N}_{\text{MPA}} \mathcal{E} \equiv -\frac{\beta_K}{2} T^{-1} \mathcal{I}^{K-1} \mathcal{E}. \quad (102)$$

### 3 Summary of Physical Model

Here we summarize the physical model that we discretize and solve in order to investigate various aspects of nonlinear optical propagation. It includes paraxial diffraction, group velocity and third-order dispersion, linear and nonlinear shock, Kerr and Raman effects and ionization. There are many models for nonlinear optical propagation in the literature [8, 9, 13, 20, 4, 18, 30, 5, 38]. Ours is similar to those in the aforementioned references. However, our method of including the impedance effects in the plasma defocusing term leads to a difference in the shock terms from those in [13].

The propagation equation for the electric field envelope  $\mathcal{E}$  is

$$\partial_\zeta \mathcal{E} = \frac{i}{2k} T^{-1} \nabla_\perp^2 \mathcal{E} + iD\mathcal{E} + T\mathcal{N}\mathcal{E}, \quad (103)$$

with wavenumber  $k = 2\pi/\lambda$  and wavelength  $\lambda$ . The dispersion and shock operators are given by

$$D \equiv -\frac{k''}{2} \partial_\tau^2 - i \frac{k'''}{6} \partial_\tau^3 \quad \text{and} \quad T \equiv \left(1 + \frac{i}{\omega_o} \partial_\tau\right) \quad (104)$$

respectively. The nonlinear operator may be decomposed as

$$\mathcal{N} = \mathcal{N}_{\text{kr}} + T^{-1} \mathcal{N}_{\text{pls}} + T^{-1} \mathcal{N}_{\text{MPA}}. \quad (105)$$

Kerr and Raman effects are modeled with

$$\mathcal{N}_{\text{kr}} = ik_o n_2 [(1 - \alpha)\mathcal{I} + \alpha \mathcal{Q}_R]. \quad (106)$$

We have used the approximately time-harmonic relationship

$$\mathcal{I} \approx \frac{cn_o \epsilon_o}{2} |\mathcal{E}|^2 \quad (107)$$

to specify the intensity envelope  $\mathcal{I}$ . Delayed Raman response is described by

$$\mathcal{Q}_R(\mathbf{r}, t, z) = \int_{-\infty}^t R(\tau) \mathcal{I}(\mathbf{r}, t - \tau, z) d\tau, \quad (108)$$

with

$$R(t) = \frac{\gamma^2 + \omega_R^2}{\omega_R} \exp(-\gamma t) \sin(\omega_R t), \quad (109)$$

where  $\omega_R$  and  $\gamma$  are respectively characteristic frequency and attenuation constants for the molecular species interacting with the pulse.

The following quantities

- $\tau_c$  = Electron collision or relaxation time
- $m_e$  = Mass of an electron
- $\omega_o$  = Pulse center frequency
- $k_o$  = Pulse center wavenumber
- $n_o$  = Background refractive index
- $K$  = Photons required to free single electron
- $U_i$  = Energy required to free single electron
- $\rho_{\text{nt}}$  = Neutral atom number density
- $\alpha_r$  = Electron-positive ion recombination loss
- $\sigma_k$  = Ionization cross-section.

help us express the remaining ionization physics. For plasma defocusing we use

$$\mathcal{N}_{\text{pls}} \mathcal{E} = -\frac{ik_o}{2n_o\rho_c} T^{-1}(\rho \mathcal{E}), \quad (110)$$

or if we desire to include collision effects

$$\mathcal{N}_{\text{pls}} \equiv -\frac{\sigma}{2}(1 + i\omega_o\tau_c)\rho, \quad (111)$$

with

$$\rho_c \equiv \epsilon_o m_e \omega_o^2 / e^2, \quad \text{and} \quad \sigma \equiv \frac{k_o}{n_o \rho_c} \frac{\omega_o \tau_c}{1 + \omega_o^2 \tau_c^2}. \quad (112)$$

The energy loss in the pulse due to multiphoton absorption is included via

$$\mathcal{N}_{\text{MPA}} = -\frac{\beta_K}{2} \mathcal{I}^{K-1}, \quad (113)$$

with

$$\beta_K = \sigma_k \rho_{\text{nt}} K \hbar \omega_o. \quad (114)$$

The evolution of the electron density requires an auxiliary equation

$$\frac{d\rho}{dt} = \frac{\sigma}{n_o^2 U_i} \rho \mathcal{I} + \mathcal{I}^K \sigma_k (\rho_{\text{nt}} - \rho) - \alpha_r \rho^2, \quad (115)$$

or

$$\frac{d\rho}{dt} = \frac{\sigma}{n_o^2 U_i} \rho \mathcal{I} + \frac{\beta_K}{K \hbar \omega_o} \mathcal{I}^K - \alpha_r \rho^2. \quad (116)$$

# A Symbols

Symbol	Units	Description
$\epsilon_o = 8.854 \times 10^{-12}$	$\text{F} \cdot \text{m}^{-1}$	Permittivity of free space
$\mu_o = 4\pi \times 10^{-7}$	$\text{H} \cdot \text{m}^{-1}$	Permeability of free space
$e = 1.60217657 \times 10^{-19}$	C	Magnitude of the charge carried by an electron
$c = 2.99792458 \times 10^8$	$\text{m} \cdot \text{s}^{-1}$	Speed of light <i>in vacuo</i>
$\mathcal{F}, \mathcal{F}^{-1}$	—	Fourier and inverse Fourier transforms
$\mathbf{E}, E, \mathcal{E}$	$\text{V} \cdot \text{m}^{-1}$	Electric field (vector, scalar, envelope)
$\hat{E}, \hat{\mathcal{E}}$	$\text{V} \cdot \text{m}^{-1}$	Electric field (frequency domain scalar and envelope)
$\mathbf{H}$	$\text{A} \cdot \text{m}^{-1}$	Magnetic field (vector)
$\mathbf{D}$	$\text{C} \cdot \text{m}^{-2}$	Electric flux density (vector)
$\mathbf{B}$	$\text{W} \cdot \text{m}^{-2}$	Magnetic flux density (vector)
$\mathbf{J}, \mathcal{J}$	$\text{A} \cdot \text{m}^{-2}$	Current density (vector, envelope)
$\rho$	$\text{C} \cdot \text{m}^{-3}$	Electric charge density
$\mathbf{P}, \mathbf{P}_l, \mathbf{P}_{nl}$	$\text{C} \cdot \text{m}^{-2}$	Vector material polarization (total, linear, nonlinear)
$\chi^{(1)}$	—	Linear electric susceptibility
$\chi^{(3)}$	—	Nonlinear electric susceptibility
$\epsilon_r$	—	Relative permittivity
$t$	s	Time coordinate variable
$\omega$	$\text{rad} \cdot \text{s}^{-1}$	Angular frequency
$N_p$	$\text{C} \cdot \text{m}^{-2}$	Total nonlinear material response
$k(\omega)$	$\text{m}^{-1}$	Wavenumber as a function of angular frequency
$\hat{N}_p$	$\text{C} \cdot \text{m}^{-2}$	Fourier transform of total nonlinear material response
$\mathbf{r}$	m	Coordinate vector ( $x, y, z$ )
$k'$	$\text{s} \cdot \text{m}^{-1}$	First order coefficient in Taylor series expansion of $k(\omega)$
$k''$	$\text{s}^2 \cdot \text{m}^{-1}$	Second order coefficient in Taylor series expansion of $k(\omega)$
$k'''$	$\text{s}^3 \cdot \text{m}^{-1}$	Third order coefficient in Taylor series expansion of $k(\omega)$
$k^{(m)}$	$\text{s}^m \cdot \text{m}^{-1}$	$m$ th order coefficient in Taylor series expansion of $k(\omega)$
$\omega_o$	$\text{rad} \cdot \text{s}^{-1}$	Pulse center frequency
$x, y$	m	Transverse spatial coordinate variables
$z$	m	Longitudinal spatial coordinate variable
$\zeta$	m	Retarded longitudinal spatial coordinate variable
$\tau$	s	Retarded temporal coordinate variable
$\kappa(\omega)$	$\text{m}^{-1}$	Sum of first two terms in Taylor series expansion of $k(\omega)$
$\hat{N}_p$	$\text{C} \cdot \text{m}^{-2}$	Fourier transform of total nonlinear material response
$\mathcal{N}_p$	$\text{C} \cdot \text{m}^{-2}$	Envelope of total nonlinear material response
$\hat{\mathcal{N}}_p$	$\text{C} \cdot \text{m}^{-2}$	Fourier transform of envelope of total nonlinear material response
$\Omega$	$\text{rad} \cdot \text{s}^{-1}$	Slow frequency variable ( $\omega - \omega_o$ )
$D(\Omega)$	$\text{rad} \cdot \text{s}^{-1}$	Taylor series expansion of $k(\omega) - \kappa(\omega)$ in slow frequency variable
$T, S$	—	Shock operators
$n_o$	—	Background refractive index of material
$k_o$	$\text{m}^{-1}$	Central wavenumber
$\mathbf{N}_p$	$\text{m}^{-1}$	Vector instantaneous nonlinear polarization response (Kerr effect)
$P$	$\text{C} \cdot \text{m}^{-2}$	Scalar instantaneous nonlinear polarization response (Kerr effect)
$\mathcal{P}$	$\text{C} \cdot \text{m}^{-2}$	Scalar envelope nonlinear polarization response (Kerr effect)
$\mathcal{I}$	$\text{W} \cdot \text{m}^{-2}$	Scalar envelope intensity
$n_2$	$\text{m}^2 \cdot \text{W}^{-1}$	Nonlinear refractive index of material
$\mathcal{N}_k$	$\text{m}^{-1}$	Envelope Kerr operator
$\mathcal{N}_{kr}$	$\text{m}^{-1}$	Envelope Kerr-Raman operator
$\mathcal{Q}_R$	$\text{W} \cdot \text{m}^{-2}$	Molecular Raman response
$\gamma$	$\text{s}^{-1}$	Molecular damping rate (Raman)

Symbol	Units	Description
$\omega_R$	$\text{rad} \cdot \text{s}^{-1}$	Molecular Raman frequency
$R(t)$	—	Molecular impulse response (Raman)
$\alpha$	—	Raman scattering contribution to Kerr-Raman terms
$\rho$	$\text{m}^{-3}$	Electron number density
$K$	—	Number of electrons required to ionize atom
$\alpha_r$	$\text{m}^3 \cdot \text{s}^{-1}$	Ionization recombination loss coefficient
$\rho_{\text{nt}}$	$\text{m}^{-3}$	Number density of neutral atoms
$\sigma_k$	$\text{W}^K \cdot \text{m}^{-2K} \cdot \text{s}^{-1}$	Ionization cross-section for $K$ -photon MPI processes
$U_i$	J	Energy required to free an electron (ionization energy)
$\hbar$	$\text{J} \cdot \text{s}$	Planck's constant (reduced)
$\sigma$	$\text{m}^2$	Avalanche ionization rate coefficient
$\rho_c$	$\text{m}^{-3}$	Critical plasma number density (plasma is opaque for $\rho > \rho_c$ )
$\beta_K$	$\text{J} \cdot \text{W}^K \cdot \text{m}^{-2K-3} \cdot \text{s}^{-1}$	Ionization rate coefficient (MPI)
$\mathbf{v}$	$\text{m} \cdot \text{s}^{-1}$	Velocity
$\mathbf{F}$	N	Force
$\mathcal{P}_{\text{pls}}$	$\text{C} \cdot \text{m}^{-2}$	Plasma current equivalent polarization
$\mathcal{N}_{\text{pls}}$	$\text{C} \cdot \text{m}^{-2}$	Plasma current equivalent polarization operator
$\tau_c$	s	Electron collision or relaxation time
$\mathbf{p}(t)$	$\text{kg} \cdot \text{m} \cdot \text{s}^{-2}$	Electron momentum
$\hat{\mathcal{P}}_{\text{pls}}$	$\text{C} \cdot \text{m}^{-2}$	Fourier transform of plasma current equivalent polarization
$W$	J	Work
$P$	$\text{J} \cdot \text{s}^{-1}$	Power
$S$	$\text{J} \cdot \text{s}^{-1} \cdot \text{m}^{-3}$	Power per unit volume
$\mathcal{S}$	$\text{J} \cdot \text{s}^{-1} \cdot \text{m}^{-3}$	Envelope power per unit volume
$W_r$	$\text{s}^{-1}$	Power per unit volume
$\mathcal{P}_{\text{MPA}}$	$\text{C} \cdot \text{m}^{-2}$	MPA equivalent polarization
$\mathcal{N}_{\text{MPA}}$	$\text{C} \cdot \text{m}^{-2}$	MPA envelope operator



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